



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>7</sup> :</b> <b>A01N 25/30, 47/38, 25/04 // (A01N 47/38, 47:30, 43:653, 43:52)</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 00/07444</b> <b>(43) International Publication Date:</b> 17 February 2000 (17.02.00)
<b>(21) International Application Number:</b> PCT/GB99/02162 <b>(22) International Filing Date:</b> 19 July 1999 (19.07.99)  <b>(30) Priority Data:</b> 9816697.8                      31 July 1998 (31.07.98)                      GB  <b>(71) Applicant (for all designated States except US):</b> HOECHST SCHERING AGREVO GMBH [DE/DE]; Miraustrasse 54, D-13509 Berlin (DE).  <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> KNEEBONE, Graham [GB/GB]; AgrEvo UK Ltd., Hauxton, Cambridge CB2 5HU (GB). PATEL, Smita [GB/GB]; AgrEvo UK Limited, Hauxton, Cambridge CB2 5HU (GB).  <b>(74) Agent:</b> SEWELL, Richard, Charles; AgrEvo UK Limited, Patent Dept., Chesterford Park, Saffron Walden, Essex CB10 1XL (GB).		<b>(81) Designated States:</b> AU, BR, CA, CN, CZ, HU, ID, IL, IN, JP, KR, KZ, MX, NO, NZ, PL, RO, RU, SI, SK, TR, UA, US, ZA, ZW, European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> LIQUID FORMULATION  <b>(57) Abstract</b> <p>The invention provides a suspo-emulsion formulation comprising first and second compatible components, the first component acting primarily as an oil emulsifier and secondarily as a particle dispersant, and the second component acting primarily as a particle dispersant and secondarily as an oil emulsifier, characterised in that: i) the first component consists of one or more compounds of formula <math>R^1(Y)_aH</math>; ii) the second component consists of one or more compounds of formula <math>[R^2(Z)_b]_cA</math>; wherein <math>R^1</math> and <math>R^2</math>, which may be the same or different, are substituted phenoxy; Y and Z, which may be the same or different, are <math>-CHR^3CHR^4-O-</math>, where <math>R^3</math> and <math>R^4</math>, which may be the same or different, are hydrogen or methyl; a is 7 to 40; b is 7 to 40; c is 1 or 2; and A is phosphate or sulfate radical, or salts thereof, wherein the concentration of the first component is greater than 70 g/litre. The invention enables the preparation of stable suspo-emulsions containing a variety of active ingredients and components and containing large quantities of oily components.</p>		

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### Liquid Formulation

This invention relates to liquid formulations and methods for their preparation. In particular, the invention relates to liquid formulations that contain an emulsions phase  
5 and a suspended solid phase. Specifically, although not exclusively, the invention concerns formulations for use in agriculture, such as those containing pesticides, e.g. herbicides, fungicides and/or insecticides, where the formulations are diluted with water by an agricultural operator prior to application to the pest or its locus.

10 Creating stable emulsions that contain at least one solid component, so-called suspo-emulsions, presents significant challenges to the formulation chemist. These challenges increase the greater the number and complexity of the components. Suspo-emulsion formulations are especially sensitive to the shear forces during manufacture of the formulation, and also during dilution and mixing of the formulation with water by  
15 the agricultural operator. These forces accelerate hetero-flocculation, caused by agglomeration of the solid particles with droplets of the emulsion phase. Hetero-flocculation causes poor storage stability and may result in blocked nozzle filters during spraying of the diluted formulation by the agricultural operator. These problems are exacerbated when formulations contain large amounts of oily components which are  
20 often required to improve translocation of the active ingredient into the plant.

The challenge for the formulation chemist is to choose the right dispersants and emulsifiers to produce a stable system. Typically this is achieved by coating the solid particles with a polymer dispersant forming a steric layer around each particle; the oil  
25 component is then emulsified into the mixture with the aid of an emulsifier. The emulsion can be thermodynamically or kinetically stabilised. Kinetically stabilised systems will eventually coalesce back to a single oil phase, typically this takes about 2 to 3 years, i.e. the duration of the shelf life of the product. The thermodynamically stable systems spontaneously form microemulsion/solubilised systems when mixed  
30 together in the correct proportions.

Thus, attempts at providing stable suspo-emulsion formulations have typically involved the use of polymer dispersants to keep the droplets and particles physically apart. However these attempts have not found general applicability, because although they  
35 can be applied to specific, simple systems, they are not applicable to more complex suspo-emulsions. It is therefore the objective of the invention to provide a suspo-emulsion formulation that overcomes the above-mentioned problems.

According to the invention there is provided a suspo-emulsion formulation comprising first and second compatible components, the first component acting primarily as an oil emulsifier and secondarily as a particle dispersant, and the second component acting  
5 primarily as a particle dispersant and secondarily as an oil emulsifier, characterised in that,

- i) the first component consists of one or more compounds of formula  $R^1(Y)_aH$ ;
- ii) the second component consists of one or more compounds of formula  $[R^2(Z)_b]_cA$ ;

10 wherein

$R^1$  and  $R^2$ , which may be the same or different, are substituted phenoxy;

Y and Z, which may be the same or different, are  $-CHR^3CHR^4-O-$ , where  $R^3$  and  $R^4$ , which may be the same or different, are hydrogen or methyl;

a is 7 to 40;

15 b is 7 to 40;

c is 1 or 2; and

A is phosphate or sulfate radical (preferably phosphate), or salts thereof,

wherein the concentration of the first component is greater than 70 g/litre.

20 The term suspo-emulsion, as known in the art, describes formulations wherein the continuous phase is water containing an emulsion and suspended particles. Emulsions can be "water-in-oil" emulsions where the oil phase is the major component surrounding aqueous particles or "oil-in-water" emulsions, where the aqueous phase is the major component. Our invention finds applicability in both systems, however it is  
25 particularly advantageous for use in water-in-oil suspo-emulsions.

Throughout this specification, unless the context requires otherwise, the word "comprise", or variations such as "comprises" or "comprising", will be understood to  
30 imply the inclusion of a stated integer or group of integers, but not the exclusion of any other integer or group of integers, including method steps.

Our invention enables the preparation of stable suspo-emulsions containing a variety of active ingredients and other components. It therefore finds more general applicability than prior art formulations. Moreover our invention enables the preparation  
35 of formulations containing much larger quantities of oily components than have hitherto

been possible. We have found that formulations prepared in accordance with our invention are able to withstand high-shear conditions commonly experienced during manufacture and application of pesticides on to crops. Comparable systems, not utilising our invention, result in hetero-flocculation. By designing a system as described  
5 above where the dispersant and emulsifier are related, our invention goes against conventional teaching in the art and provides the formulation chemist with a new technical teaching which will find general applicability.

We have found that good performance is achieved when the amount of second  
10 component is between 15 to 50 g/litre, especially 25 and 35 g/litre.

We have found that our invention works particularly well when the formulation contains an ionic emulsifier. Salts of alkyl benzene sulfonates are preferred, especially, calcium dodecyl benzene sulfonate.

15

Surprisingly we have found that suspo-emulsions when prepared in accordance with the invention form emulsions wherein the water/oil interface cannot easily be distinguished. These so-called microemulsions are characteristic of our invention particularly when the total concentration of the first component and the ionic emulsifier  
20 exceeds 150 g/litre.

We have found that enhanced performance is achieved when  $R^1$  and/or  $R^2$  are substituted by two or three 1-phenylethyl groups.

25 Furthermore, we have found that a and b, which may be the same or different, are preferably from 10 to 25, especially from 16 to 20.

The invention also includes formulations wherein the second component is a salt, preferably a sodium, potassium or triethanolamine salt. Especially preferred is the  
30 triethanolamine salt.

It is particularly preferred that the first component comprises a compound of formula  $R^1(Y)_aH$ , where  $R^1$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Y is  $-CH_2CH_2-O-$  and a is nominally 16. In the art this

compound is known as ethoxylated tristyrylphenol (16 mole E.O.). Typically, this compound exists as a mixture with similar compounds.

It is particularly preferred that the second component comprises the triethanolamine salt of a compound of formula  $[R^2(Z)_b]_cA$ , where  $R^2$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Z is  $-\text{CH}_2\text{CH}_2-\text{O}-$ , b is nominally 20, c is 1 and A is phosphate. In the art this compound is known as a ethoxylated tristyrylphenol phosphate (20 mole E.O.). Typically, this compound exists as a mixture with similar compounds, particularly with similar compounds where c is 2.

10

Our invention finds application particularly in the technical field of pesticide formulation, where many active ingredients exhibit low solubility in both aqueous and organic solvents. Such active ingredients include fluquinconazole, carbendazim and isoproturon.

15

Our invention finds particular applicability in the preparation of formulations comprising two or more active ingredients, especially where one ingredient is a high melting point, water insoluble solid and another ingredient is dissolved in a non water-miscible liquid such as rape oil or mineral oil, e.g. prochloraz.

20

### Examples

The invention will now be shown, by way of example only, with reference to the following Examples.

25

### Example 1

#### Choice of non-ionic emulsifier and dispersant on formulation stability.

Formulations 1, 2 and 3 were prepared according to the following method.

### Preparation

30

Ingredients 1 to 5 were mixed in the proportions shown in Table 1 to form a solution. Ingredients 6 to 12 were mixed in the proportions shown in Table 1 and ground in a bead-mill until the fluquinconazole had a median diameter of 2 microns. Both sets of ingredients were then mixed to produce the resulting formulation. As can be seen in Table 1, the ingredients of each formulation remained constant except for ingredients 4 and 10.

35

Compound I is of formula  $R^1(Y)_aH$ , where  $R^1$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Y is  $-CH_2CH_2-O-$  and a is nominally 16. In the art this compound is known as a ethoxylated tristyrylphenol (16 mole E.O.). Typically, this compound exists as a mixture with similar compounds.

5

Compound of formula II is the triethanolamine salt of a compound of formula  $[R^2(Z)_b]_cA$ , where  $R^2$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Z is  $-CH_2CH_2-O-$ , b is nominally 20, c is 1 and A is phosphate. In the art this compound is known as a ethoxylated tristyrylphenol phosphate (20 mole E.O.). Typically, this compound exists as a mixture with similar compounds, particularly with similar compounds where c is 2.

10

#### Test Method

The stability of the formulations was tested using a laboratory scale spray rig that was designed to simulate field conditions. Each diluted formulation was re-circulated vigorously for at least half an hour using a high power circulation pump to simulate the shear forces that would be experienced in the spray tank in practice. A pre-weighed filter is positioned in an arm at right angles to the re-circulation flow and the flow through the filter is regulated so as to encourage deposition of any residue.

20

Stability of the formulations was assessed in a number of ways. A formulation was classified as having failed if after re-circulation at least one of the following applied:

- i) the weight of residue on the filter was greater than 0.02% of the original weight of formulation used,
- ii) there was a large build-up on the walls and floor of the beaker,
- iii) there was a sticky tide mark on the walls of the beaker,
- iv) microscopic evaluation revealed hetero-flocculation had occurred.

25

All four criteria were considered in determining how a formulation had performed. The test results are shown in Table 1.

30

Table 1

	Ingredient	mass/ g	Formulation 1	Formulation 2	Formulation 3
1	Prochloraz	174	-	-	-
2	Penetrant	174	-	-	-
3	Aromatic solvent	190	-	-	-
4	Non-ionic Emulsifier	215	Compound I	Compound I	Castor oil ethoxylate (40 mole EO)
5	Ionic Emulsifier	92	-	-	-
6	Fluquinconazole	54	-	-	-
7	Antifreeze	66	-	-	-
8	Antifoam	2	-	-	-
9	Crystallisation inhibitor	38	-	-	-
10	Dispersant	30	Compound II	Oleyl methyl tauride	Compound II
11	Biocide	2	-	-	-
12	Water	make up to 1 litre	-	-	-
	<b>TEST RESULTS</b>		<b>PASS</b>	<b>FAIL</b>	<b>FAIL</b>

The results show that formulations made in accordance with the invention are resistant to hetero-flocculation.



Example 2

Effect of concentration of Compound I (non-ionic emulsifier) and Compound II (dispersant) on formulation stability.

- Formulations 4 to 10 were prepared according to the preparative method described in Example 1 in proportions shown in Table 2. The proportions are shown in grams. Table 2 also shows the test results which were assessed using the same criteria as in Example 1.

Table 2

	<b>Ingredient</b>	<b>4</b>	<b>5</b>	<b>6</b>	<b>7</b>	<b>8</b>	<b>9</b>	<b>10</b>
1	Prochloraz	174	174	174	174	174	174	174
2	Penetrant	174	174	174	174	174	174	174
3	Aromatic solvent	190	190	190	190	190	190	190
4	Compound I	70	105	122.5	126	140	175	215
5	Ionic Emulsifier	30	45	52.5	54	60	75	92
6	Fluquinconazole	54	54	54	54	54	54	54
7	Antifreeze	66	66	66	66	66	66	66
8	Antifoam	2	2	2	2	2	2	2
9	Crystallisation inhibitor	38	38	38	38	38	38	38
10	Compound II	25	25	25	25	25	25	25
11	Biocide	2	2	2	2	2	2	2
12	Water (make up to 1 litre)							
	<b>TEST RESULTS</b>	<b>FAIL</b>	<b>PASS</b>	<b>PASS</b>	<b>PASS</b>	<b>PASS</b>	<b>PASS</b>	<b>PASS</b>

- 10 The results show that hetero-flocculation occurs when the amount of compound I falls below 70 g/litre.

CLAIMS

- 1 A suspo-emulsion formulation comprising first and second compatible  
5 components, the first component acting primarily as an oil emulsifier and  
secondarily as a particle dispersant, and the second component acting primarily  
as a particle dispersant and secondarily as an oil emulsifier, characterised in  
that,
- 10 i) the first component consists of one or more compounds of formula  
 $R^1(Y)_aH$ ;  
ii) the second component consists of one or more compounds of formula  
 $[R^2(Z)_b]_cA$ ;
- wherein  
 $R^1$  and  $R^2$ , which may be the same or different, are substituted phenoxy;  
15 Y and Z, which may be the same or different, are  $-CHR^3CHR^4-O-$ , where  $R^3$   
and  $R^4$ , which may be the same or different, are hydrogen or methyl;  
a is 7 to 40;  
b is 7 to 40;  
c is 1 or 2; and  
20 A is phosphate or sulfate radical, or salts thereof,  
wherein the concentration of the first component is greater than 70 g/litre.
- 2 A formulation according to claim 1 wherein the amount of second component is  
between 15 to 50 g/litre.
- 25 3 A formulation according to claim 1 wherein the amount of second component is  
between 25 and 35 g/litre.
- 4 A formulation according to claims 1 to 3 which further contains an ionic  
30 emulsifier.
- 5 A formulation according to claim 4 wherein the ionic emulsifier is a salt of an  
alkyl benzene sulfonate.

- 6 A formulation according to claims 4 or 5 which is a micro-emulsion wherein the total concentration of the first component and the ionic emulsifier exceeds 150 g/litre.
- 5 7 A formulation according to any preceding claim wherein  $R^1$  and/or  $R^2$  are substituted by two or three 1-phenylethyl groups.
- 8 A formulation according to any preceding claim wherein a and b, which may be the same or different, are from 10 to 25.
- 10 9 A formulation according to any preceding claim wherein a and b, which may be the same or different, are from 16 to 20.
- 15 10 A formulation according to any preceding claim wherein A is phosphate.
- 11 A formulation according to any preceding claim wherein the second component is a sodium, potassium or triethanolamine salt.
- 12 A formulation according to any preceding claim wherein the first component comprises a compound of formula  $R^1(Y)_aH$ , where  $R^1$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Y is  $-CH_2CH_2-O-$  and a is nominally 16.
- 20 13 A formulation according to any preceding claim wherein the second component comprises the triethanolamine salt of a compound of formula  $[R^2(Z)_b]_cA$ , where  $R^2$  is tri-substituted phenoxy, substituted by a 1-phenylethyl radical at the 2,4 and 6 positions, Z is  $-CH_2CH_2-O-$ , b is nominally 20, c is 1 and A is phosphate.
- 25 14 A formulation according to any preceding claim which contains at least one active ingredient exhibiting low solubility in both aqueous and organic solvents.
- 30

# INTERNATIONAL SEARCH REPORT

Inter-Office Application No.

PC1/GB 99/02162

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 A01N25/30 A01N47/38 A01N25/04 //(A01N47/38, 47:30, 43:653, 43:52)

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 A01N

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## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 297 207 A (HOECHST AG ;BAYER AG (DE)) 4 January 1989 (1989-01-04) see: page 2, line 25 - 29; compounds I - III; cpds VI and VII; cpd. IX; page 6, line 23 - page 7, line 15; page 8, line 5 - 8; example 10. ---	6-14
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☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

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